

Appln No. 10/087,247

Amdt date November 25, 2003

Reply to Office action of August 25, 2003

Amendments to the Claims:

This listing of claims will replace all prior versions, and listings, of claims in the application:

Listing of Claims:

1. (Original) A carbonaceous material comprising:
a graphite particle having a 002 plane interval d002 of less than 0.337 nm, as measured by the X-ray wide angle diffraction method;
a complex particle disposed and distributed in the vicinity of the surface of the graphite particle, the complex particle comprising silicon and carbon and having a particle size smaller than that of the graphite particle; and
an amorphous carbon layer having a 002 plane interval d002 of more than 0.37 nm, as measured by the X-ray wide angle diffraction method, the amorphous carbon layer being a polymer layer and being coated on the graphite particle and the complex particle rendering them bound;
wherein the complex particle comprises a Si particulate, a conductive carbon material disposed and distributed in the vicinity of the surface of the Si particulate, and a rigid carbon material layer coated on the Si particulate and the conductive carbon material rendering them bound, the Si particulate being composed of a crystalline Si phase.
2. (Original) The carbonaceous material according to claim 1, wherein the crystalline Si phase is deposited with at least

Appln No. 10/087,247

Amdt date November 25, 2003

Reply to Office action of August 25, 2003

one phase selected from SiO₂ phases, SiC phases, and SiB₄ phases.

3. (Original) The carbonaceous material according to claim 1, wherein the silicon and the carbon are present in a weight ratio of 0.1:99.9 to 50:50.

4. (Original) The carbonaceous material according to claim 2, wherein the P_{SiO₂}/P_{Si} ratio is no less than 0.005 and no more than 0.1 and the P_{SiC}/P_{Si} ratio is no less than 0.005 and no more than 0.1, wherein P_{Si} is defined as the diffraction intensity of the plane (111) of the Si phase, P_{SiO₂} is defined as the diffraction intensity of the plane (111) of the SiO₂ phase, and P_{SiC} is defined as the diffraction intensity of the plane (111) of the SiC phase, measured by the X-ray wide angle diffraction method.

5. (Original) The carbonaceous material according to claim 2, wherein the P_{SiO₂}/P_{Si} ratio is no less than 0.005 and no more than 0.1, the P_{SiC}/P_{Si} ratio is no less than 0.005 and no more than 0.1, the P_{SiB}/P_{SiO₂} ratio is no less than 0.1 and no more than 5.0, and a P_{SiB}/P_{SiC} ratio is no less than 0.1 and no more than 5.0, wherein P_{Si} is defined as the diffraction intensity of the plane (111) of the Si phase, P_{SiO₂} is defined as the diffraction intensity of the plane (111) of the SiO₂ phase, P_{SiC} is defined as the diffraction intensity of the plane (111) of the SiC phase, and P_{SiB} is defined as the diffraction intensity of the plane (104) of the SiB₄ phase, as measured by the X-ray wide angle diffraction method.

Appln No. 10/087,247

Amdt date November 25, 2003

Reply to Office action of August 25, 2003

6. (Currently Amended) The carbonaceous material according to Claim 1, wherein the graphite particle has a particle size ranging from ~~2 to 70 μm~~ , 2 to 70 μm the complex particle has a particle size of no less than 50 nm and no more than 2 μm , and the amorphous carbon layer has a thickness of no less than 50 nm and no more than 5 μm .

7. (Original) The carbonaceous material according to claim 1, wherein the Si particulate has a particle size of no less than 10 nm and less than 2 μm , the conductive carbon material has a specific resistance of no more than $10^{-4} \Omega \cdot \text{m}$, and the rigid carbon layer has a flexibility strength of no less than 500 kg/cm² and a thickness of no less than 10 nm and no more than 1 μm .

8. (Original) The carbonaceous material according to claim 1, wherein the complex particle is present in an amount no less than 1% by weight and no more than 25% by weight.

9. (Original) The carbonaceous material according to claim 1, wherein the amorphous carbon layer is obtained by heat-treating at least one polymer material selected from the group consisting of thermoplastic resins, thermosetting resins, vinyl-based resins, cellulose-based resins, phenol-based resins, coal-based pitch materials, petroleum-based pitch materials, and tar-based materials.

10. (Original) The carbonaceous material according to Claim 9, wherein the mixing weight ratio of Si : graphite : polymer is 0.1:99.8:0.1 to 40:40:20.

Appln No. 10/087,247

Amdt date November 25, 2003

Reply to Office action of August 25, 2003

11. (Original) A lithium secondary battery comprising the carbonaceous material according to Claim 1.

12. (Currently Amended) A method of preparing a carbonaceous material comprising the steps of:

calcining a Si particulate composed of a crystalline Si phase in a carbon crucible at ~~1300 to 1400 °C~~ 1300 to 1400 °C to deposit a SiO₂ phase and a SiC phase in the crystalline Si phase;

adding a conductive carbon material to the Si particulate;
applying a polymer material coating solution to the Si particulate to provide a complex particle precursor;

calcining the complex particle precursor to render the polymer material coating solution into a rigid carbon layer to provide a complex particle;

adding the complex particle to a graphite particle;

applying a polymer material coating solution to the graphite particle to provide a carbonaceous material precursor;
and

calcining the carbonaceous material precursor to render the polymer material coating solution into an amorphous carbon layer to provide a carbonaceous material.

13. (Currently Amended) A method of preparing a carbonaceous material comprising the steps of:

calcining a Si particulate together with a B₂O₃ powder in a carbon crucible at ~~1300 to 1400 °C~~ 1300 to 1400 °C to deposit SiO₂, SiC, and SiB₄ phases in a crystalline Si phase;

adding a conductive carbon material to the Si particulate;

Appln No. 10/087,247

Amdt date November 25, 2003

Reply to Office action of August 25, 2003

applying a polymer material coating solution to the Si particulate to provide a complex particle precursor;

calcining the complex particle precursor to render the polymer material coating solution into a rigid carbon layer to provide a complex particle;

adding the complex particle to a graphite particle;
applying a polymer material coating solution to the graphite particle to provide a carbonaceous material precursor; and
calcining the carbonaceous material precursor to render the polymer material coating solution into an amorphous carbon layer to provide a carbonaceous material.